LOADING AND TESTING A LIGHT SCATTERING CELL WITH A BINARY FLUID MIXTURE NEAR ITS CRITICAL COMPOSITION

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FINAL REPORT

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Table of Contents

Turbidity Cell Filling with Methanol Cyclohexane

PART I. INTRODUCTION

It is impossible to expect that an experimental endeavor could simply begin and continue without a process of preparation or initiation, or that a thought could just mature without some history linking the past to the present. The fact is that the only way to succeed in the experimental sciences is through forward progress. Progress that takes into account the many failures and successes of the past and keeps in mind also the hope of the future. It is this lifetime process of change and adaptation that attracts so many to the field of experimental physics. It is this same natural law that guides the field of critical phenomena and turbidity. Critical phenomena has been the subject of physics research for many years now. However, only in recent years has the research effort become intense. The current intensity has caused the study of critical phenomena to be grouped into a previous older era and a present contemporary era.(Stanley, 1971) *Turbidity Cell Filling with Methanol Cyclohexane* is one of the first steps toward a further understanding of critical phenomena. The research period, outlined in the following report and manual, was spent developing apparatus and techniques that will make it possible to study critical phenomena through turbidity measurements on methanol cyclohexane. Topics covered in this manual range from the orientation of turbidity cell parts for assembly to the filling apparatus and procedure used when the cell is built. The last section will briefly cover some of the observations made when viewing the cell in a controlled water bath. However, before mention is made of the specifics of the summer research, a short introduction to critical phenomena and turbidity and how they relate to this experiment is needed.

Critical Phenomena and Turbidity

The special aspect of critical phenomena is the universal way to describe many systems near their critical point. The universality doesn't refer to the behavior of the system in terms of its components, but each system is related with regards to its critical properties. A process of scaling a system by its critical point helps describe these critical properties universally. Consider for example, a system of two fluids with closely matched densities acting as a homogeneous mixture because the temperature of the sample is over its critical point. As the temperature begins to decrease and get closer to the critical temperature, droplets begin to form as the fluids begin to go to a two phase system. When these droplets become big enough to be seen, the system takes on a cloudy appearance. This cloudy appearance is called critical opalescence. In defining the size of these droplets as they grow, an average length is given to them, or correlation length ξ. This correlation length is related to the critical properties by,

$$\xi = \xi_0 t^{-\upsilon}$$

where $t \equiv (T - T_c)/T_c$ is the reduced temperature, ξ_0 is the amplitude describing the correlation length far from the critical temperature T_c, and υ is a universal critical exponent approximately equal to 0.63. Other equations involving critical exponents include

$$\chi = \chi_o t^{-\gamma}, \text{ T>T}_\text{C}$$

and

$$\phi - \phi_c = Bt^{\beta}, \text{ T<T}_\text{C}$$

where χ is the osmotic compressibility, χ_o is the osmotic compressibility amplitude, t is the reduced temperature, ϕ is the volume fraction of one component in the upper or lower phase, ϕ_c is the critical composition, B is the amplitude of the coexistence curve,

and γ and β are critical exponents. The critical exponents are the part of binary systems that are universal. Universal here is not used freely. Universal means that no matter what system in a universality class that is investigated, the values for these exponents do not change.

One exponent that has not been studied as much as the others is η. η, which is an exponent used to describe light scattering at small angles, has yet to be confirmed through experiment. Evaluating this exponent with intent to get a concrete determination is the eventual goal that *Turbidity Cell Filling with Methanol Cyclohexane* is initializing. Turbidity is the pathway to be used to study critical phenomena and methanol cyclohexane is the binary fluid mixture that will provide the environment. However far off the goal of studying the critical phenomena behind methanol cyclohexane, the procedure of assembling and filling a cell with the critical composition of methanol cyclohexane is an essential link to the future of physics research in this ever expanding field.

Turbidity Cell Filling with Methanol Cyclohexane spanned a ten week research period during the summer of 1992, where James Becker (co-author of this report) worked under Dr. D.T. Jacobs on the campus of The College of Wooster, Wooster, Ohio and has been continued during the subsequent academic year by Anne Flewelling, also under Jacobs' direction. The information recorded in this manual is a synopsis of what occurred.

PART II. TURBIDITY CELL PART ORIENTATION FOR ASSEMBLY

The assembly of the Turbidity Cell needs to be done with careful consideration to the following directions. The process of assembling, filling, sealing, and studying the cell involves it being attached or involved with a variety of different apparatus. There are extra holes on the primary cell clamps, what we will call 'flanges,' to allow the cell to be secured to the different apparatus. However, in order for the securing of the cell to be possible, it is crucial that the cell be assembled correctly.

It is understood that the cell being assembled may be of two different kinds. The only actual difference between the cells is the differing flanges. One set is made of aluminum and has a flattened side on it. The other set is made of copper and is round. The reason for the difference is irrelevant, but the process of putting the two kinds together may represent some difficulty. The assembly instructions below have taken into account the existence of this peculiarity and will provide proper direction for both types of cells.

Turbidity Cell Part Cleaning Procedure

Cleanliness is an important aspect of Turbidity Cell filling. The process of cleaning the items needs to be followed carefully, as there are many different causes for impurities. An impurity can be anything from a large scratch on the surface of the window, thus distorting vision, to a fingerprint on the knife edge that causes the cell to leak. Impurities can also be dirt or debris trapped in the cell that alter the critical composition of the fluid entered into the cell. Throughout the instructions, when mention is made to clean according to the "instruction given previously," use the following procedure.

1. Ultrasonic Clean
   a. Place the item(s) to be cleaned in a beaker 3/4 full of distilled water. The beaker should be big enough to fit in the ultrasonic cleaning bath.
   b. Put a small amount of MICRO liquid laboratory soap in the beaker. (MICRO liquid laboratory cleaner made for critical cleaning, manufactured by International Products Corp., Trenton, NJ.)
   c. Put the beaker in the ultrasonic cleaning bath for 3 to 5 minutes.

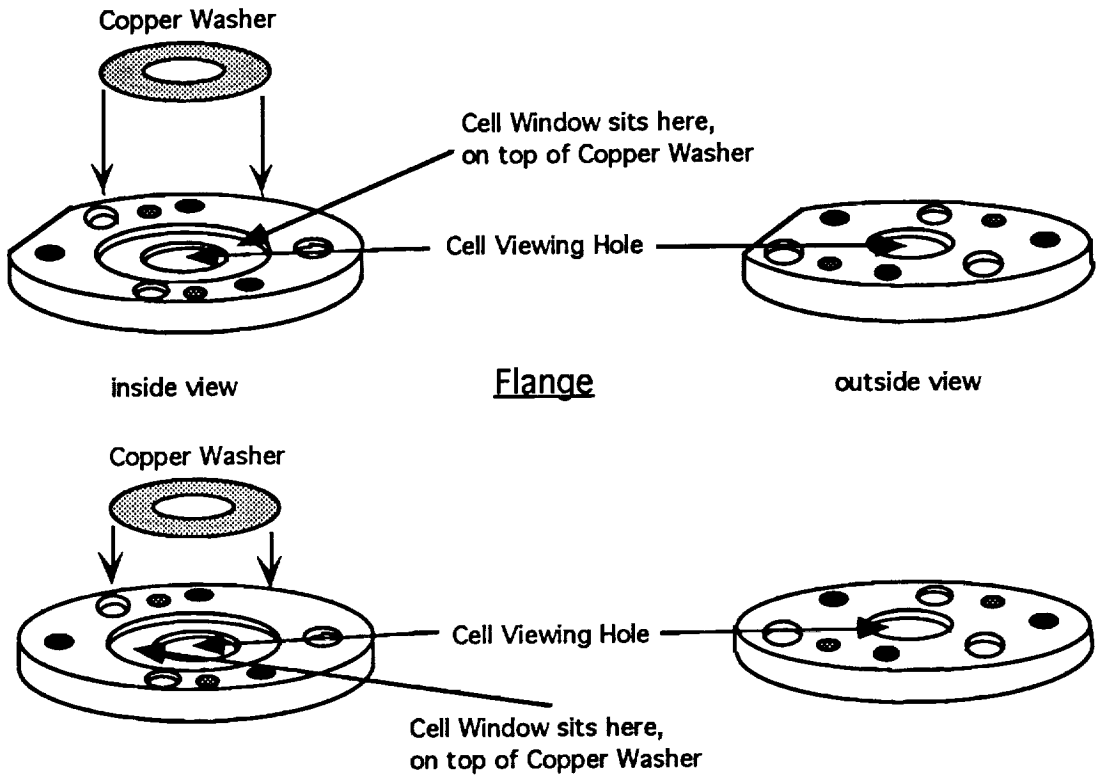
d. Remove and rinse with distilled water and acetone.

2. Acid Clean
   a. Put enough of a dilute acid (H_2SO_4) in a small beaker to cover the item(s). Make sure the acid is not too strong.
   b. Put the item(s) in the acid with plastic tweezers and wearing gloves. It is a good idea to use the tweezers and wear gloves when working with acid and also because a fingerprint can cause a bad seal, possibly leading to a leaky cell.
   c. Allow the item(s) to sit in the acid for 3 to 5 minutes. If, at any time during the acid cleaning, bubbles form on the outside of the item(s) remove it immediately and rinse it off with distilled water. This means that the acid is too strong as it was reacting with the metal. Add distilled water to the acid solution to dilute it more and put the item(s) back in.
   d. After 3 to 5 minutes has passed, remove the item(s) with the tweezers and rinse it thoroughly with distilled water.
   e. Inspect the item(s) for dirt, oxidation, or other blemishes careful not to touch items that can't be handled (This will be explained later) and repeat full procedure as needed.
3. If the item(s) seems clean then rinse again with distilled water and then with acetone. Put out on paper towel to air dry, then put in Al foil and place in vacuum oven (60°C, –20 psi).

Turbidity Cell Part Description

The next step is to get acquainted with the different components of the cells so that the instructions may be written with some clarity and understanding. The labels that have been placed near the objects, naming certain parts of these objects, are used throughout the instructions and should be read carefully so that general knowledge of the cell is acquired.

1. Flange: The flanges are the part of the cell that provide rigidity. They structure the cell by holding it together. Each flange has a hole through it, called the Cell Viewing Hole, for viewing the contents of the cell after it has been filled. Notice that the hole on one side of the flange is bigger, that is the place where the Cell Window (see below) will sit. It faces in, toward the actual cell. The screws that hold the flanges together are called Support Screws and are machine size 6-32. Six of these screws provide the pressure to seal the cell. The manner in which the support screws connect the flanges is discussed at the end of this section. Two smaller screw holes are used to secure the cell to different apparatus. They are called the Secure Screw Holes and are machine size 4-40. The top two flanges pictured below are aluminum, notice the flat side, and the bottom two round flanges are copper. The left two are showing the inside view. Inside view means that the inside of the cell is nearer that side of the flange. In other words, the cell windows sit in the sunken portion of the flange, and the sunken portion can be seen by viewing the inside of the flange. A Copper Washer sits in the sunken portion between the flange and the cell window to protect the window from breaking. Copper is a soft metal and will bend and flatten to protect the glass window. All flanges are identical with regards to the cell viewing hole and other holes. All have the sunken portion so that the windows fit "in" the flanges. The two flanges on the right are showing the outside view, meaning that side of the flange is farther away from the cell than the inside view. There are two flanges per cell.



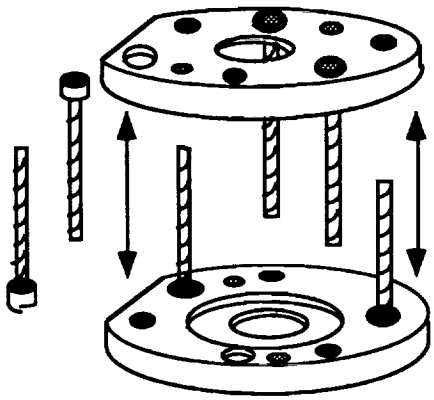
The following drawing simply illustrates how the support screws fit into the flanges. Each flange has six support screw holes. Three of these are female and are threaded able to accommodate the support screws. The other type of hole doesn't have threads and only allows the screw through the flange to thread into the other flange. This type of hole shall be referred to as a countersink style hole. When the flanges are put together, the screws will go through the countersink type of hole in the one flange and thread into the female hole of the other flange. On the drawing a dark circle is a female hole with threads and the countersink style hole is represented by an opening in the flange. Of course, the drawing below on the right is meaningless since the cell itself is not inserted. However, a good way to get use to the workings of the flanges and support screws, is to practice without the windows in. In regards to operations involving support screws, the copper flanges are identical to the aluminum ones.

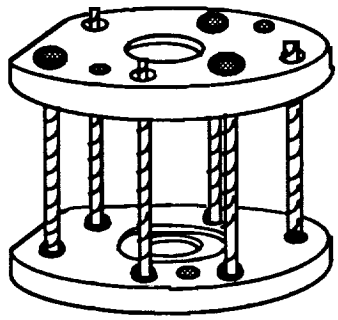
= countersink style hole
with hanging screw =

= female receptacle style hole

= secure screw hole

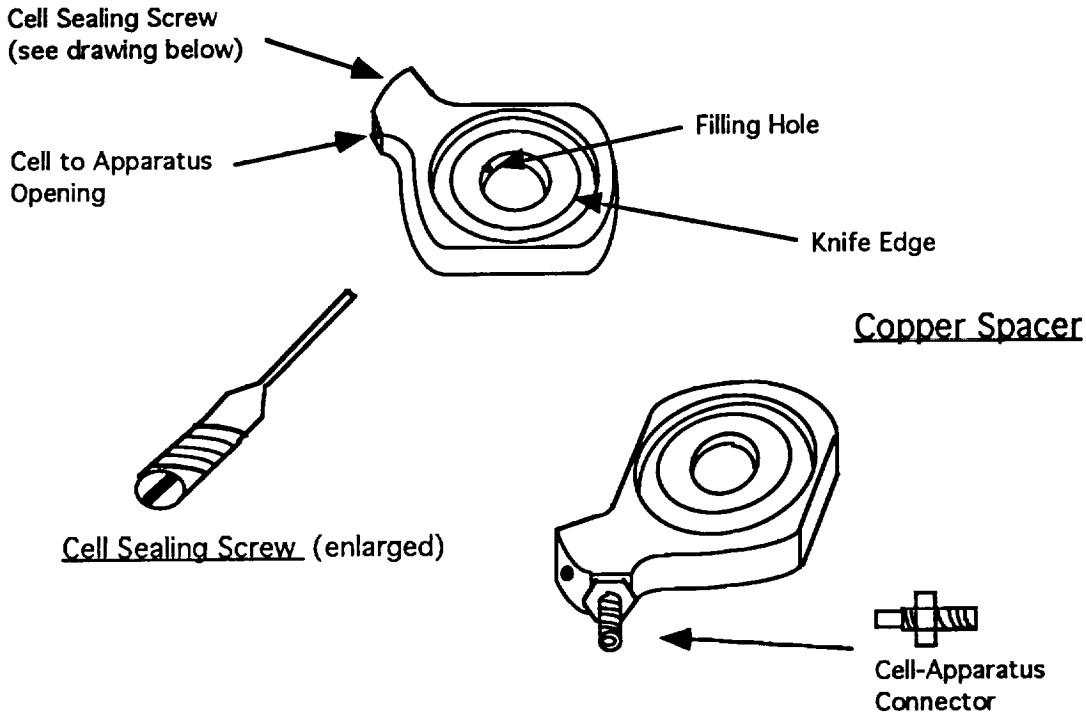
Support Screw and Flange Workings



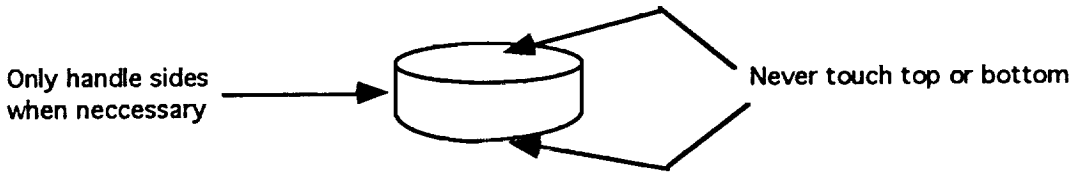


2. Copper Spacer: The copper spacer forms the cell because it separates the Cell Windows (see below) and provides an opening for the filling of the cell using the cell to apparatus opening. The apparatus is connected by means of the cell-apparatus connector, which requires a 5/16" open-end wrench and soldering to attach. A problem with the spacer and the cell-apparatus connector that has occurred again and again, is the lack of a firm seal to fill and empty the cell. Therefore, the actual cell assembly instructions that follow illustrate the machining, fitting, and soldering that needs to be done to seal this connection.

After the cell is filled, by means of the filling hole, the Cell Sealing Screw in the spacer is tightened to seal its contents. The cell sealing screw is brass. A knife edge on the spacer provides the necessary seal to form the cell between the two cell windows. There is one spacer per cell.



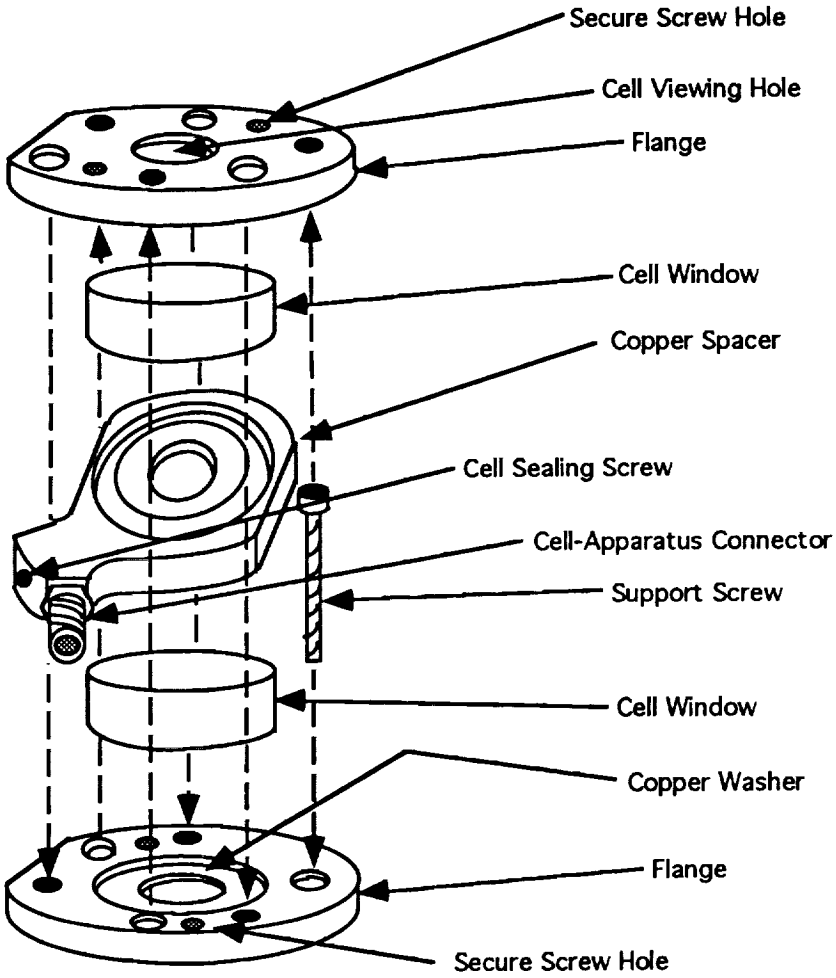
3. Cell Windows: The cell windows are made of BK-7 glass and are separated by the spacer and form the cell. Never touch the top or bottom of the windows as this may damage them and impair the quality with which the filled cell may be observed. Only touch the sides of the windows if need be and only handle them when about to use. Otherwise, let them sit in the protective case they came in. There are two windows per cell.



Cell Window

General Part Orientation

Now that a general idea of what the parts of the cell look like has been achieved it is possible to begin learning the orientation of the parts to each other as the cell is being assembled. Below is a drawing of the orientation of the parts to each other. Notice, first, how the flanges are oriented. (they are the aluminum set) The cell windows fit into the bigger hole in the one side of each flange. Thus, with both cell windows centered, the copper spacer fits between the two.



The aluminum flanges have only one orientation to each other when the inside is correctly fit with the windows. The flat sides on the flanges must be parallel with each other. Notice that when the flanges are set together, with the flat sides parallel, the secure screw holes do not match up. The copper flanges do not have a flat side to match up and so they can be turned such a way that they do have matching secure screw holes. The matching of the secure screws holes from flange to flange is not important. What is important is that the support screw holes line up and even more important is the orientation of the cell-apparatus opening to the support screws, with respect to the location of the secure screw holes.

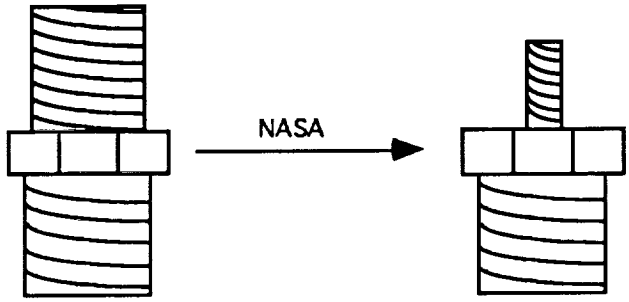
If support screws of different lengths are available, then carefully put the cell together like the above illustration. Don't use the cell windows, but imagine they are in place as well. Choose the support screws that will thread through one flange into the other and not stick outside of the flange. They will be between 6-32-3/4 and 6-32-1, depending on the thickness of the spacer and window used. The support screws provided by NASA are titanium. We had Invar screws constructed to minimize the thermal expansion differential between the screws and the glass windows.

Turbidity Cell Assembly

In order to correctly assemble the cell:

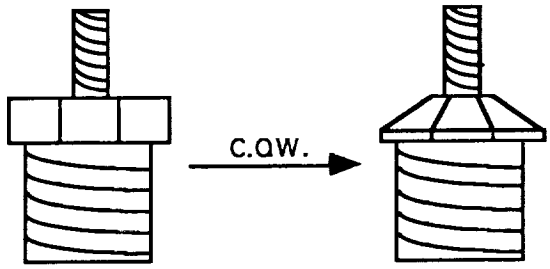
1. Wash both flanges, both copper washers, the spacer, and the cell sealing screw following the cleaning procedure mentioned before.
2. Special Preparation of the spacer and the cell-apparatus connector.

The history behind the creation of an effective cell-apparatus connector begins with a Swagelok® piece called a male connector, catalog number B-100-1-1. Half of this piece was altered so one side would thread into the spacer (machine size 4-40) and the other side, still supporting a 5/16" hex, joined the cell to the filling apparatus.



Male Connector: B-100-1-1 Cell-Apparatus Connector

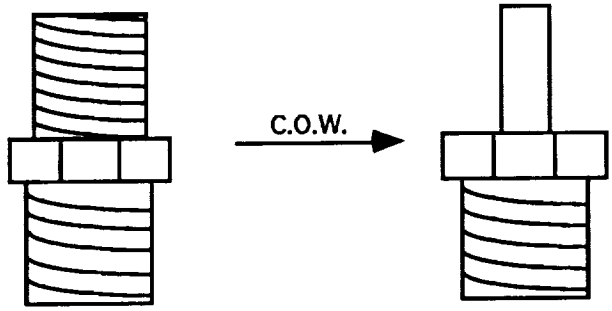
The cell-apparatus connector was not effective for a few reasons. First, altering the male connector meant that the sides of the altered portion were very weak. Secondly, when it was trimmed to a 4-40 size, the length of the threads was also shortened. Thus, when the c-a connector was threaded in, the hex on the c-a connector would hit a support screw and put unneeded sideways pressure on the weak threads. The first solution to this problem was to push the spacer to one side as far as it would go, hopefully leaving enough room between the hex and the support screw to allow a good connection. However, even though the connector did go in farther it still resulted in a bad seal and a broken thread. This problem must have been a sum result of the weak threads and the unneeded sideways pressure applied to the threads. The next step was to grind off some of the hex as to alleviate the possibility of any sideways pressure applied to the c-a connector.



Cell-Apparatus Connector Modified C-A Connector

To solve the problem with the weak threads the decision was made to solder the c-a connector to the spacer. Soldering the pieces, when grouped with the grinding down of the hex, should have provided a stronger, superior seal. The modified c-a connector was threaded into the spacer and soldered. Soldering threads is possible, however, the seal that is formed is not as secure as a seal formed by two flush surfaces. When the cell was being reattached to the apparatus for emptying, the solder didn't hold when the slightest bit of torque was applied to the c-a connector. If soldering is the answer to a good seal, then the threads have to be removed so a more solderable surface is formed. Because soldering seemed the only answer to a strong enough seal and the threads would not solder well, the only option was to remove the threads. However, removing the threads from the already weak c-a connector would not accomplish a stronger connection at all. The only option was to order a male connector, the piece from which alterations led to the

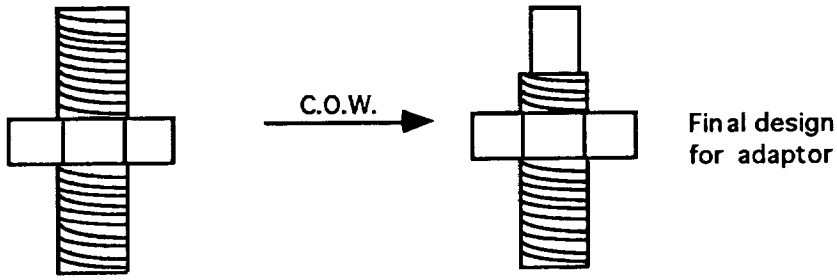
original c-a connector, and to lathe off the threads. This would leave a round, smooth end that could be inserted into the spacer and soldered.



Male Connector: B-100-1-1 Modified Male Connector

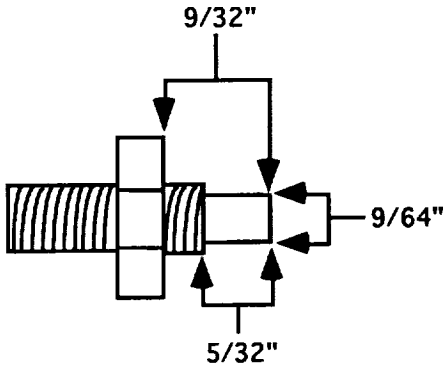
The hole in the spacer would also have to be drilled out to accommodate for a bigger c-a connector, but the result would be a joint that contains two pieces that are flush. This connection worked very well. It turned out that because the male connector had a longer end on it, the hex wouldn't hit one of the support screws, and consequently not apply a sideways pressure on the c-a connector.

The only remaining problem with the c-a connector did not lie in the cell to apparatus connection, it was between the c-a connector and the next piece, a female connector, Swagelok® catalog number B-100-7-1. The female connector joined the c-a connector with the 1/16" tubing that was part of the apparatus. Attaching and detaching the cell many times wore on the 1/16" tubing and caused it to break right near the female connector. The solution was relatively straight forward. Now that the c-a connector was self-manufactured, the problem could be solved by ordering a male connector that would accept the 1/16" tubing on one side and be altered for cell connection on the other side, thus getting rid of the need for the female connector all together. This turned out to be a union, Swagelok® catalog number B-100-6.



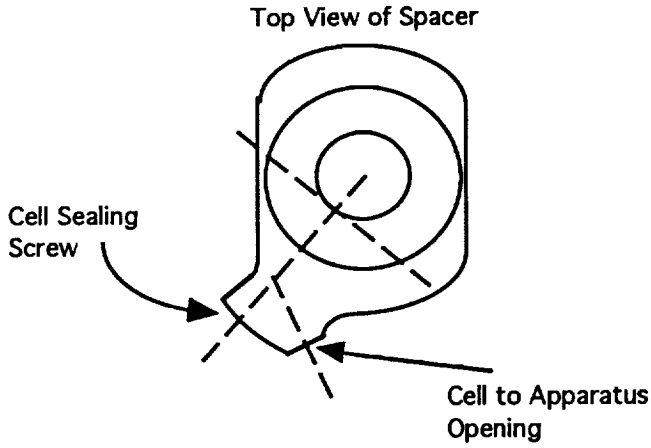
Union: B-100-1-1 Modified Union

a. The threads of the union need to be removed. It is advantageous to leave as much of the cylinder beneath the threads as possible. This will insure a sturdy connection with the spacer. Using a metal lathe, remove the threads from one side of the union. The threads are 9/32" long, but not all of them need to be removed. Removing about 5/32" (a little more than 1/8) is sufficient. If only the threads are removed from the union, the diameter of the cylinder will be about 9/64".



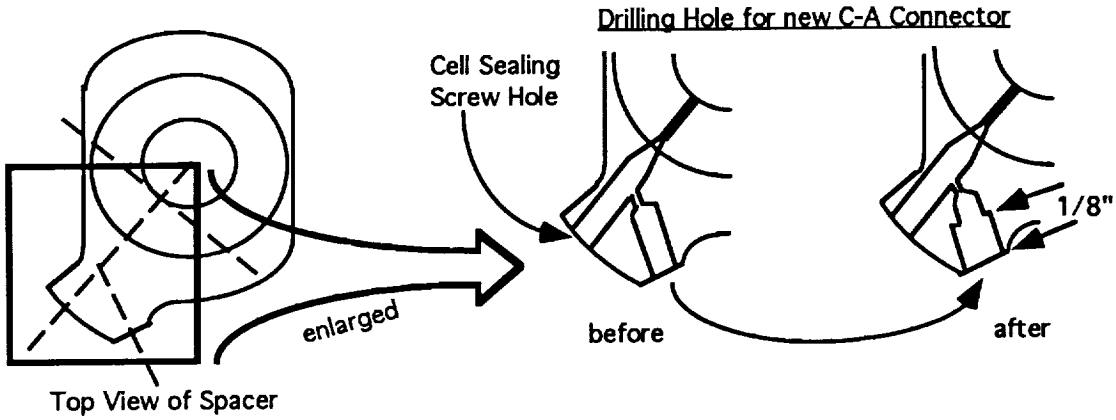
Modified Union (C-A Connector) Measurements

b. The cell to apparatus opening in the spacer needs to be drilled out. There are two holes in the spacer, one is for the cell sealing screw and one is for the filling apparatus. Make sure the correct hole is drilled out. The hole for the cell sealing screw is perpendicular to a tangent to the hole in the middle of the spacer. That is why the cell sealing screw is straight. It screws into and fills the filling hole to seal the cell. The hole for the apparatus isn't as symmetrical. It joins the channel for the cell sealing screw at an angle. The angled hole is the one to be drilled.

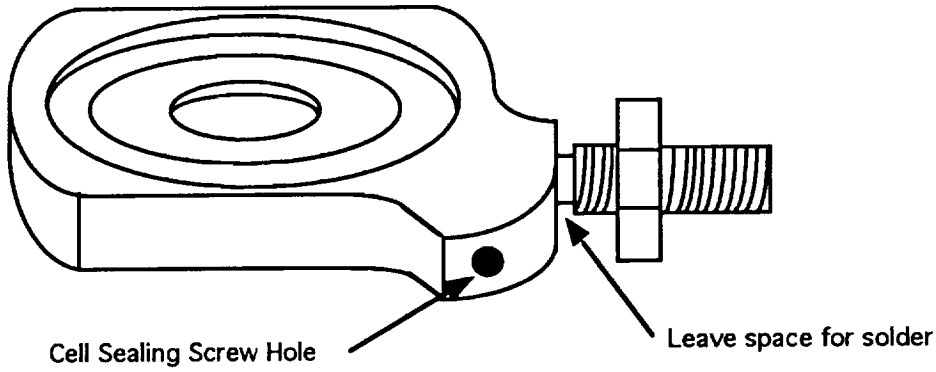


If you lathed the union to the measurements above, then a #28 drill bit (.140", ~9/64") should be used to drill the hole. If you measure something different then use a different drill bit. An accurate device for measuring these is a dial caliper.

The drill bit should be centered in the cell to apparatus opening of the spacer so that it is straight, lined up with the existing channel. Make sure the spacer is square so the bit doesn't wander off to the side and leave a crooked hole. Only drill the hole 1/8" deep. If you lathed off 5/32" of threads, then when the modified union (c-a connector) is put in the hole, a small amount (~1/32") of the lathed part will stick out. This is good because it will allow the solder a place to sit and insure that the lathed end is all the way down in the hole. Thus, when the soldering is done, there is a smaller chance for the solder to fill the hole you just drilled. If the connector doesn't touch the bottom of the spacer hole, the solder may flow under the connector and fill the hole.



c. Place the new c-a connector in the newly drilled hole and see if it fits. If adjustments need to be made, make sure as much of the cylinder on the union is left intact. This will help with the strength of the connection. The new c-a connector should not fit too loosely in the spacer or be too tight, although it helps if it is straight. The picture below shows a close up of the new c-a connector in the spacer. Note the small amount of lathed cylinder that protrudes from the hole, allowing the solder to sit and seal the joint and insuring that the cylinder touches the bottom of the drilled hole.



d. Wash both items as described previously to clear the joint of oxidation.
e. After the parts are dry, apply flux to them carefully. A few hints when applying the flux is that if the flux fills the hole, the solder will fill the hole. Apply the flux only to those parts you wish to be joined and don't apply too much. Make sure there is some flux on the cylinder portion of the c-a connector and at the top of the joint where there is a small space before the threads. We used Harris stay-clean flux and stay-brite 1/30 silver-tin solder.
f. Put the pieces together and place them down on the surface to be heated. When heating the pieces, keep in mind that copper conducts heat very well and thus the joint will heat up quickly. Heat the joint indirectly. Do not let the flame touch the joint and keep the flame away from the inside of the spacer as much as possible. Heating the knife edge may cause it to harden or may cause the top of a thin knife edge to burn away. If it is at all possible, only brush the flame near the outside of the spacer and the far end of the c-a connector. Remember, any oxidation that is on the spacer will have to be cleaned before assembly. While heating, check to see if the joint is hot enough by placing the solder on it. If the joint is too hot then the solder will not stick. Use only a small amount of solder. Using too much, even though it seems to be going on the joint, probably means that the solder is filling the hole. A good technique to use when solder the pieces is placing

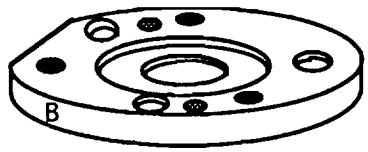
the solder on the joint with some protruding over the other side. If some solder protrudes over the other side, it will wrap around the joint, thus ridding the need to flip the spacer over and possibly damage the newly soldered joint. When the soldering is done, allow ample time for the pieces to cool.

g. After the piece is cool, squirt distilled water in the holes to make sure they are open and allow fluid to pass. Thread the cell sealing screw into its appropriate hole to make sure it is working, then remove it so it doesn't break. If water can flow through the c-a connector into the spacer then go on to the next step. If it seems that the hole is plugged, then drill the solder out of the spacer/c-a connector with a very small drill bit. The smallest opening in the union is .05 inches in diameter, so the recommendation is to use a drill bit smaller than that.

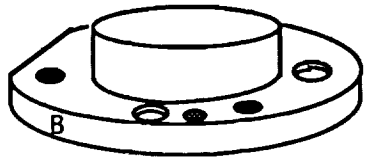
h. Clean the spacer/c-a connector piece thoroughly as previously mentioned.

i. No matter how much caution was used when heating the copper spacer for soldering and no matter how thoroughly the pieces were cleaned it is likely that oxidation still remains on the spacer. This will need to be cleaned off with very fine steel wool (000 size). Wet the spacer first and use laboratory soap with the steel wool. Only rub the spacer in circular motion. It is helpful to hold the steel wool still and rotate the spacer on it. Using a small portion of the steel wool will help clean the indentation in the spacer, as well as the areas near the knife edge. Rotating the spacer will avoid making radial scratches on the knife edge which ruin the spacer's sealing capabilities. When all of the oxidation appears to be removed from the spacer, rinse it with distilled water and clean again following the special procedure on the first page.
The spacer should now be ready for assembly

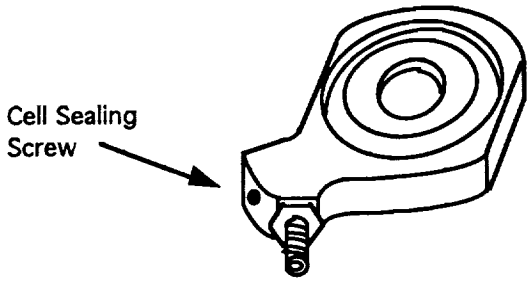
3. Place an aluminum flange on the table so that the inside may be seen. A cell window will fit in it. Align the flat side so that it is on the left. If the flange is round, align so that the holes are identical to the drawing below. Put a copper washer in the flange. This flange should be marked somehow (a paint pen is sufficient). Keeping track of what flange was initially on the bottom during assembly will help when mounting the cell on the aluminum plate for filling.



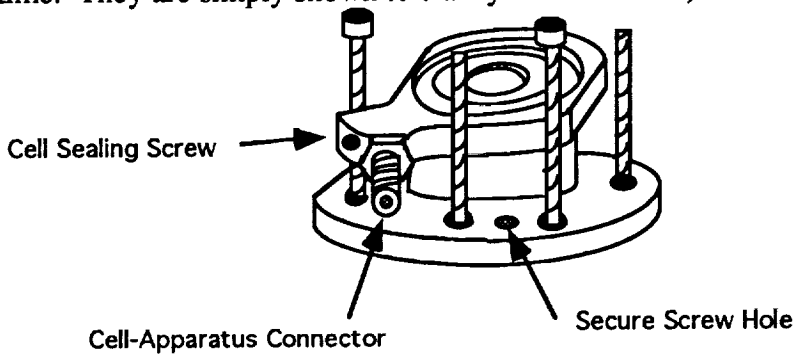
4. Take one of the cell windows and clean it off with methanol. Moisten an optical lens wipe with methanol and gently wipe the window on both sides. When one side is wiped hold it up to the light, vertically, and ensure that it dries evenly. If any dirt is accumulated on the window surface, the methanol will not dry as quickly near that point. If dirt is on the window surface, continue cleaning until it dries evenly. This procedure must be done for both sides. One side of the window has an anti-reflection coating on it, giving a blue tint when light is reflected off of it. Hold the window up to the light to figure out which side is treated. The treated side of the window is placed toward the outside of the cell, away from the spacer, and touching the copper washer on the flange. Place the first cell window into the bottom flange, with the coated side down.



5. Placement of the spacer is the most important step in assembling the cell. When it is put on top of the cell window, the cell sealing screw must be to the left and the cell to apparatus opening is to the right. The portion of the spacer that contains these two openings must be positioned as shown below, almost on the left side as you view it.



Place the spacer on the first cell window so that it is between the two support screws just in front of the flat side. The cell to apparatus opening on the spacer will be near a countersink style hole with the screw running up and threading into the top flange. The cell sealing screw will be near a female threaded hole in the flange on the table. (The screws should NOT be in place at this time. They are simply shown to clarify the directions)



This orientation would be the same for both flanges. The cell-apparatus connector is placed near a screw that may either thread in the top or in the bottom flange, but what is important is where the secure screw holes are. On the other side of the support screw nearest the cell-apparatus connector must be a secure screw hole, just as in all of the drawings as of yet. If the spacer openings (cell-apparatus opening, cell sealing screw) can be seen between to support screws straight on, then the secure screw hole must be off to the right of the right support screw. As long as a secure screw hole is off to the right of the right support screw on the bottom flange while looking straight at the cell-apparatus connector it is assembled correctly.

6. Clean the next cell window the same way as the first and place it on the spacer. Makes sure the specially coated side is up, or toward the outside of the cell.

7. Carefully place the other copper washer on the top of the cell window. It needs to be centered as much as possible, but avoid scratching the window.

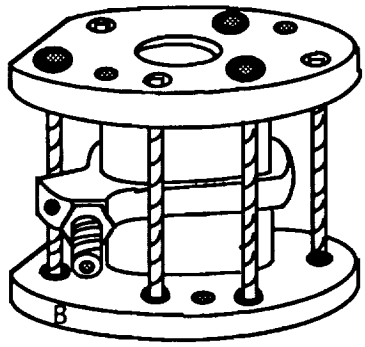
8. Align the top flange so the support screw holes match up with the bottom flange. Of course "match up" means that countersink style holes are across from female threaded holes and vice versa. The aluminum flanges are easy to match up. The flat sides have to be flush with each other. Once again, if you mark and remember which flange is on the bottom, then the alignment of the secure screw holes from flange to flange is not important, although proper alignment of the support screw holes is essential.

9. Hold one support screw up to the side of the cell and make sure the correct length is being used. When put in the holes, the screws should not stick out the other side, but they should thread into the flange.

   Put the support screws through the countersink style holes in the top flange until they meet the female holes of the second flange. Screw them in slowly, until the head of each screw is flush with the top flange. Do not put any extra torque on the screws, as this may press down the copper knife edge of the spacer. Make sure the screws appear to be flat and even. Don't have one tighter than the others so that the flange is crooked. Carefully pick up the cell and turn it over. Do the same for the screws when the bottom flange is now on top. If the cell has aluminum flanges with a flat side then make sure that they are flush. This can be checked by placing the cell on a table and using the flat surface of the table to align the flanges. No matter what type of flange, make sure the spacer is properly aligned. This may involved rotating the spacer some.

10. Using a sensitive torque wrench tighten each of the screws on one side 1/8 of turn and then turn the screws on the other side 1/8 of a turn. Turning the screws a small amount one after another, insures a flush seal with an evenly crushed copper knife edge. Continue this until a force of 2 inch-pounds is being applied to the cell. If dealing with aluminum flanges, make sure the flat sides are flush.

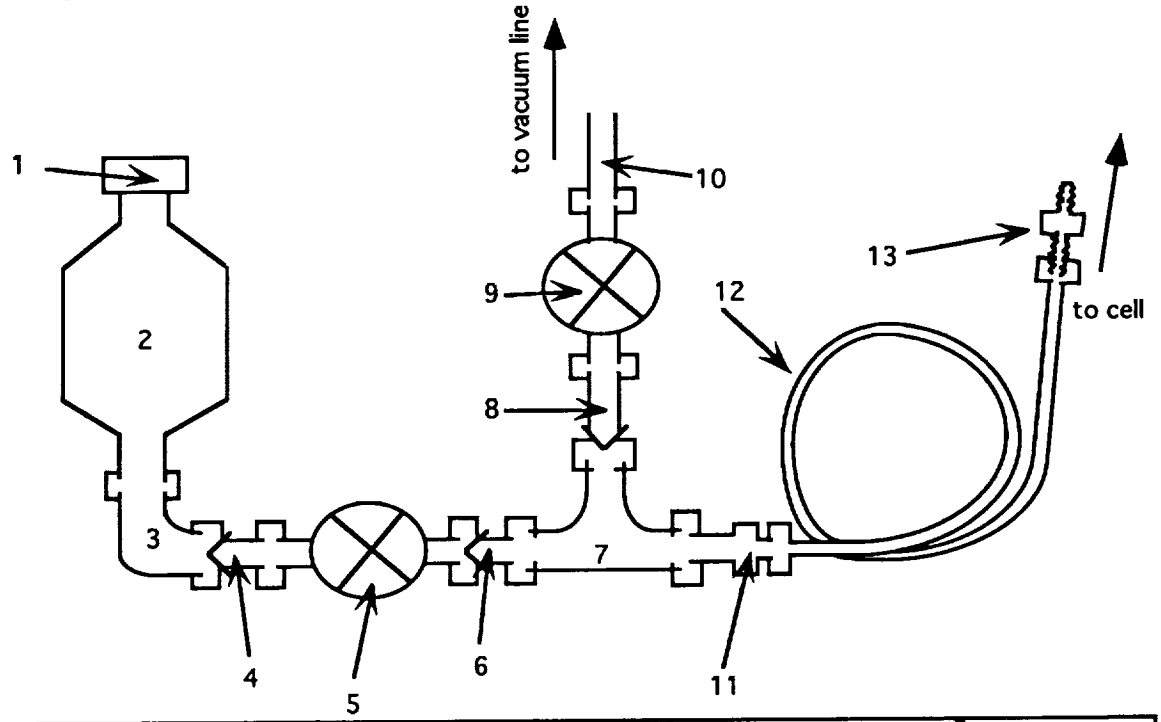
11. Repeat step 11 until a force of 5 inch-pounds is applied to the support screws.



PART III. TURBIDITY CELL FILLING APPARATUS

The apparatus used to fill the turbidity cell whose assembly was just described consists of a series of Swagelok® pieces mounted to an aluminum plate. Thermofoil heaters on the aluminum plate provide the heat required to transfer the methanol cyclohexane, in one phase, to the cell. The following paragraphs and diagrams describe the machining of the plate, placement of the Thermofoil heaters, and the different Swagelok® pieces used in the apparatus.

The center of the apparatus is a union tee so that one branch of the tee is the vacuum system, one is the cylinder of m ethanol cyclohexane, and one is the cell. Valves allow the cell to be evacuated without affecting the cylinder of fluid. When a vacuum is created in the cell, manipulating the valves will force the fluid into the vacuum, thus filling the void in the cell. The diagram below shows the apparatus with the name and Swagelok® catalog number of each piece.



No.	Name and Swagelok® ID	No.	Name and Swagelok® ID
1	cap : SS-600-C	8	port connector : SS-401-PC
2	10 cc cylinder : SS-4CD-TW-10	9	valve : SS-4BK
3	elbow : SS-600-9	10	1/4" copper tubing
4	3/8 -> 1/4 reducing port connector : SS-601-PC-4	11	1/4 -> 1/16 reducer : SS-100-R-4
5	valve : SS-4BMG	12	1/16" stainless steel tubing
6	port connector : SS-401-PC	13	modified union (c-a connector) is soldered to cell : B-100-6
7	union tee : SS-400-3		

Specific Swagelok® installation instructions accompany the pieces upon delivery.

The aluminum plate is 12.5" long, 9" wide, and 1/4" thick. Seven holes were drilled through the plate to secure the apparatus. 2 of the holes were to secure the valves, 2 to secure the cell, 1 to view the cell, and 2 to secure the cylinder. The diagram below shows the location of the holes in the plate with respect to the apparatus. The majority of the apparatus is underneath the plate. The only exception to this is the valve handles which protrude through the holes cut for them. One valve hole, for the valve near the cylinder, is cut like a slot so the cylinder portion of the apparatus can be removed and replaced. This makes it easier to fill the cylinder without dismantling the entire apparatus.(see pgs. 6-9) The three holes that are in line with each other are for the cell. The secure screw holes on the cell's bottom flange line up with the two small holes. The middle hole is for viewing the cell when it is attached to the apparatus. The other two small holes hold the cylinder against the plate by means of a small copper strap.(see pg. 8)

The diagram below also shows the elements used to heat the plate and apparatus for transferal of the methanol cyclohexane at 50°C. The two Minco Thermofoil heaters are held on the bottom of the plate by self-adhering elastic tape. The heaters are hooked up in parallel so that the resistance of the system is 180 Ohms. In parallel, the voltage needed isn't as high as in series because in series the resistance would have been 720 Ohms. Initially, a Kepco power source provided the voltage to the heaters. The plate was heated without the apparatus at 10 volts and then at 20 volts. The heat is transferred throughout the plate by conduction. Encompassing the apparatus and plate with bubble wrap will diminish the loss of heat by convection. If it is known that,

$$Power = \frac{V^2}{R} = \frac{dQ}{dt} = \mathrm{K}(T - T_R)$$

and conduction means that,

$$\frac{dQ}{dt} = \frac{\kappa A_c}{\ell}(T - T_R)$$

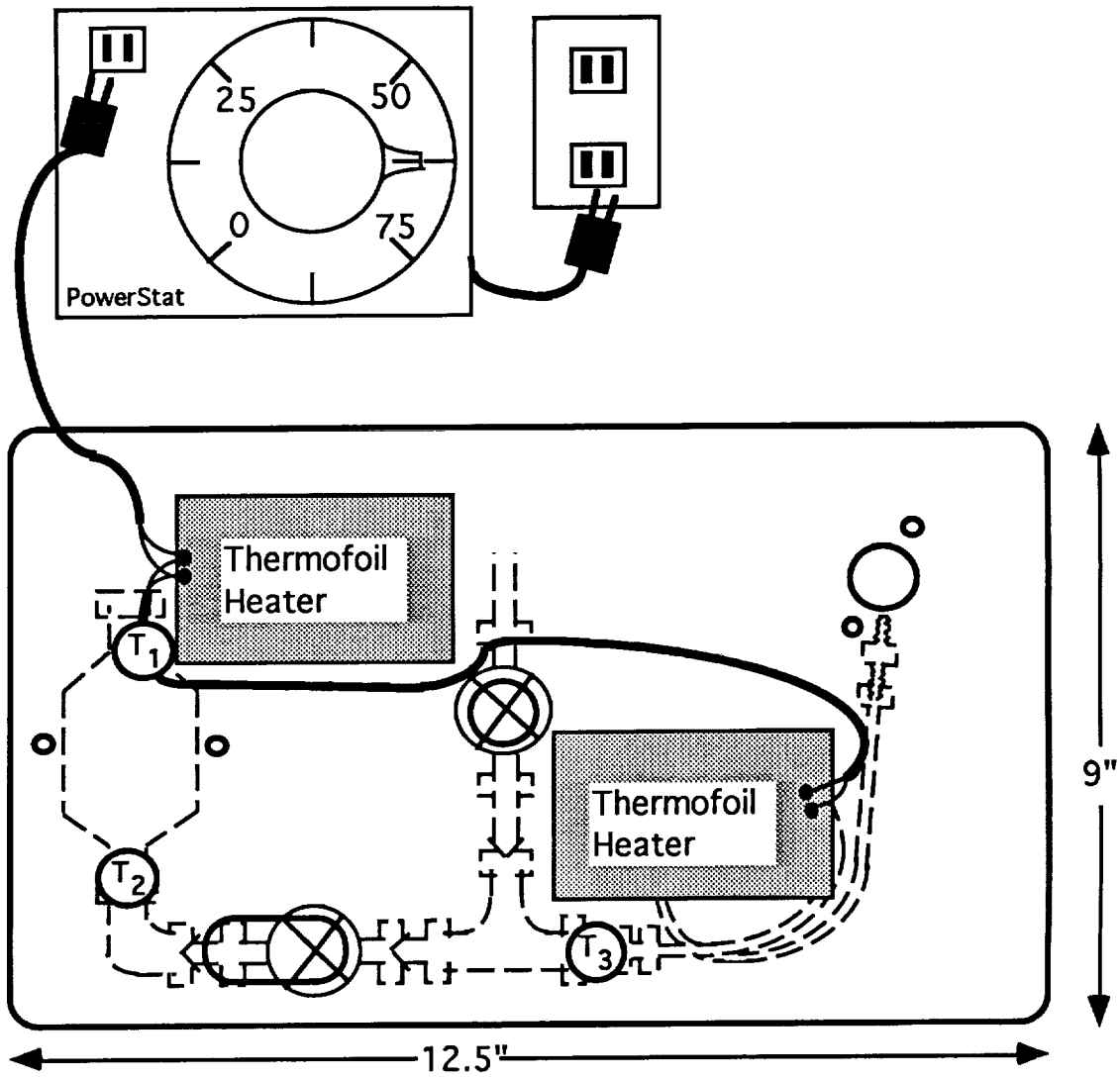
then,

$$\frac{dQ}{dt} = \frac{\kappa A_c}{\ell}(T - T_R) = \frac{V^2}{R}$$

and,

$$T = T_R + \left(\frac{\ell}{\kappa A_c R}\right)V^2$$

So, $\frac{(T - T_R)}{V^2}$ is constant with regards to heat loss in this system. Therefore, by taking the temperature attained when the plate was heated by 10 and then 20 volts, I was able to extrapolate and find out that to heat the plate to 50° Celsius, approximately 63 volts would be needed. Unfortunately, when the apparatus was added to the plate, 63 volts did not heat the entire apparatus up to 50 degrees. To heat the plate and apparatus to 50 degrees, about 82 volts is used. Instead of hooking up many Kepco power sources in series, a Superior Electric Company Powerstat "Variac" is used. A large dial on the face of the powerstat controls the AC voltage directly from the wall. Gradations on the dial are percentages of the maximum output voltage. Therefore, 100% is about 120 volts. The temperature of the plate and apparatus is measured by three thermistors. They are symbolized below as circles with a T and their number subscripted inside. The first thermistor is imbedded in the epoxy that holds the heater's wire to the plate. This thermistor registers a higher temperature than the other thermistors, which are attached to the apparatus with Apiezon Sealing Compound. See the legend at the bottom of the diagram.



Legend

T_1, T_2, and T_3 = YSI 44004 Precision Thermistor
(Resistance = 2252 Ohms @ 25° C)

The resistance of the each thermistor is read by a Keithley Multimeter.

The Minco Thermofoil Heaters are specified at a 360 Ohm resistance.

Dial gradations on the PowerStat are percentages of wall voltage.
(100% = ~120 volts)

PART IV. TURBIDITY CELL FILLING PROCEDURE

The filling of the turbidity cell is an intermediate step in the study of the critical phenomena that can only be done following preparation of the cell and apparatus. The actual filling procedure consists of heating the plate and apparatus above 50°C so the binary fluid is in one phase when it is maneuvered into the cell. However, attempts at filling the cell are futile if no caution was used when preparing the cylinder of fluid or preparing the apparatus for the filling.. This section of the manual is an attempt to relay some hints about the process of turbidity cell filling that will help avoid problems in the future. It must be understood that the following techniques were discovered by the rigorous and frustrating trial and error method.

Cleaning the Apparatus

Clean, as is suggested in the subtitle above, not only means free of dirt and debris, but it means free of other impurities such as residue from water or acetone. Therefore, all activities dealing with the cylinder used for filling the cell need to be done in a dry box. The nitrogen atmosphere of a dry box will insure that water is not present in the cylinder along with the sample of the fluid. Water not only has an adverse affect on the critical point of a binary fluid, it can also create changes in the critical composition and critical exponents.(Tveekrem and Jacobs, 1983) Other impurities can also create problems. Acetone has been proven to have created shifts in the critical parameters as well.(Cohn and Jacobs, 1984) However, simply placing the items in a dry box does not insure that they are clean. In order to properly clean the apparatus, follow these instructions;

1. Remove the portion of the apparatus from the cylinder valve to the cylinder. This part of the apparatus is called the cylinder portion. It includes everything from the valve near the cylinder (hence the name cylinder valve) to the cylinder, including the cap.. Detach the Swagelok® connection just beyond the valve, on the union tee side. The slot hole was cut so this portion of the apparatus could be detached and removed.
2. Place the cylinder and valve in a vacuum oven at ~60°C and -20 psi for 3 or more hours. Make sure the valve is open and the cap is off the cylinder. This will insure that the cylinder is dry enough to put in the dry box for fluid transfer.
3. Remove the hot cylinder portion from the oven and place it in the dry box to cool. If it was allowed to cool outside of the dry box that would defeat the purpose of drying it in the first place, as moisture would collect on it from the air. Also place a sample of methanol, a syringe, 2 crescent wrenches that are the right size to attach the cap to the cylinder, and a digital scale in the dry box.
4. When the apparatus is cool, record its weight with the Mettler digital balance in the dry box. Close the cylinder valve. Fill the cylinder with methanol keeping the cylinder valve closed so the methanol is trapped in the cylinder . Do the fluid transfer using a syringe when the cylinder cell is on the scale so before and after weight measurements can estimate the amount of fluids entered into the cylinder. The cylinder only holds 10 cc.
5. Cap the cylinder.
6. Remove the cylinder portion from the dry box and attach it to the rest of the apparatus.
7. Put the apparatus in a vacuum oven at ~60°C and -20 psi for 5 or more hours.
8. Remove the apparatus and carefully open the cylinder valve which will force the hot methanol through the apparatus, cleaning it. Only open the valve in an

area with ample ventilation and have the vacuum valve opened when the cylinder valve is opened so the copper pipe that goes to the vacuum line is cleaned.

9. Remove the cylinder portion of the apparatus once again and place it in the dry box. Close the cylinder valve, remove the cap, and fill it one more time with methanol. Cap the cylinder, remove it from the dry box, and attach it to the apparatus.
10. Heat the apparatus again at ~60°C and -20 psi for 5 or more hours in a vacuum oven.
11. When the apparatus is heated, remove it from the vacuum oven and open the cylinder valve again, only this time, leave the vacuum valve closed so the fluid is forced out of the small 1/16" tubing, thus cleaning it.

When the cleaning process is done, take the cylinder portion off again and empty the remaining methanol from the cylinder.

Preparing the Cylinder

To prepare the cylinder for a cell filling, detach the cylinder portion of the apparatus and dry it in the vacuum oven as was done before, cap off and valve open. Place it in the dry box to cool. Accompanying the cylinder portion of the apparatus to the dry box should be 2 syringes, a beaker, the samples of methanol and cyclohexane, the crescent wrenches, and the scale. The methanol and cyclohexane used in the summer of 1992 were 99 + % pure and were sealed by the Aldrich Sure Seal System.

The methanol cyclohexane that is added to the cylinder needs to be as close to the critical composition as possible. A critical composition of methanol cyclohexane is 28% methanol by weight. The bottles of fluids tell the density of the fluids on the outside. The density of the methyl alcohol was .791 g/cm^3. The density of the cyclohexane was .779 g/cm^3. A good amount of methanol cyclohexane to put in the cylinder is 9 cc. It holds 10 cc and there is a very small amount held in the apparatus elbow, but overflowing the cylinder just heightens the chance for error in mixing a critical composition. If the total mixture is 7 grams then,

$$7(.28) = 1.96 \text{ g} = 1.96 \text{ g } (\text{cc}/.791 \text{ g}) = 2.48 \text{ cc methanol}$$

and

$$7 \text{ g} - 1.96 \text{ g} = 5.04 \text{ g} = (\text{cc}/.779 \text{ g}) = 6.47 \text{ cc cyclohexane},$$

for a total of 7 grams and 8.95 cc.

When filling the cylinder, make sure the cylinder is on the scale and situated such that it can be read while filling is occurring. Make sure the valve is closed. If the methanol is being put in the cylinder first and, accidentally, too much is put in, then simply recalculate how much cyclohexane is needed for the new amount of methanol. Then be careful when adding the cyclohexane as the process of adjusting the amounts of fluids can become a never-ending circle. Put excess fluids in the beaker, not back in their bottles. Keep the bottles closed when not in use.

After the fluids are in the cylinder, put the cap on it. Record the weight of the cylinder with the cap on so that it is easier to tell how much fluid is left in the cylinder after a filling attempt. A single filling attempt doesn't take much fluid at all, ~2.5 cc, and so a full cylinder can be used for quite a few fillings. Weighing the cylinder and estimating the amount of methanol cyclohexane remaining is the best way to tell if enough fluid remains.

Remove the cylinder portion from the dry box and attach the cylinder to the apparatus. The cylinder valve should remain closed.

Attaching Cell to Apparatus

The next step is to attach the cell to the apparatus. The unmodified portion of the union (c-a connector) connects directly to the 1/16" tubing. However, when attaching the tubing to the cell, much care needs to be taken to avoid breaking the newly soldered c-a connector away from the cell. It is a good idea to gently put a pair of vise-grip pliers on the hex of the c-a connector so all of the torque is transferred to the pliers instead of being applied to the joint. Do not attach the cell to the plate until the tubing is attached. This will give the cell some flexibility in case a force is applied on it. Holding the cell near the plate will insure that the cell is flush with the plate so that it will still attach after the tubing is connected. Make sure the correct flange is near the plate when the tubing is being attached. If the cell was assembled correctly, the bottom flange during assembly is the proper flange to be against the plate. After the cell is connected to the tubing, carefully attach the cell to the plate with 4-40 screws. If the cell needs to be moved slightly before it can be attached to the plate, do not bend the tubing by holding the cell. Bend the tubing, holding the tubing, to align the cell's secure screw holes over the holes that were drilled.

Attachment of Cell Sealing Screw

Attachment of the cell sealing screw is important because if it is attached wrong, then it is hard to pull a vacuum on the cell. To be prepared for filling, the cell sealing screw needs to be wrapped in Teflon tape and threaded into its appropriate hole until only 1 thread shows outside of the spacer. This will allow fluid to pass by the end of the screw into the cell, but will not allow fluid or air to pass outside of the cell.

The Filling

The filling of the turbidity cell implies two things. First, that a turbidity cell has been assembled following the orientation instructions starting on page 3. The second implication is that the filling apparatus has been constructed, including;

1. The cleaning of the apparatus following the procedure above,
2. The mounting of the apparatus and the Thermofoil heaters to the aluminum plate,
3. The attachment of the cell to the apparatus and to the plate,
4. The cell sealing screw is in place,
5. The preparation of the thermistors for temperature measurements, (see the section on *Cell Filling Apparatus* above for reference)
6. The attachment of the vacuum to its appropriate place on the apparatus,
7. The acquisition of plastic bubble wrap to slow heat loss by encompassing the plate and apparatus, and
8. Have a small bag of crushed ice should be nearby, but still in refrigeration.

The filling process takes about 2 hours after the above preparatory steps have been taken. While the plate and apparatus are heating up, it is a good idea to keep a record of what temperature or resistance the thermistors are. This will insure that the temperature is rising at a reasonable rate. If, at any time during the heating, the temperature seems to suddenly heat slower, the bubble wrap is probably not covering the whole apparatus. Make sure there are no places for air to flow by the apparatus through the bubble wrap. Covering the apparatus includes covering the valve handles that protrude through the plate. The cylinder valve should be closed and the vacuum valve should be open.

The vacuum needs to be run for 60 - 120 minutes during heating and before fluid transfer. The cell used in this experiment is made to withstand the pressures of liquid-gas

mixtures and so pumping a vacuum for longer than 120 minutes only helps the fill. Pumping for less time only increases the chance of not getting enough fluid to the cell. The vacuum line has a cold trap.

Trial and error have reduced the process of filling to an experiment in timing. The goal is for the apparatus to reach 50°C at the same time the vacuum has been pumping for ~120 minutes. Then it is simply a matter of a few steps to fill the cell:

1. Near the end of the filling process, the apparatus will have to be shaken to insure the fluids in the cylinder are in one phase with each other. This is simply a matter of grabbing the apparatus and tilting it back and forth to shake up the fluids inside the cylinder. At this time, don't worry about keeping the apparatus covered with bubble wrap. The small amount of time in the air won't drop the temperature enough to force the fluids into two phase. Shaking should take between 1 and 2 minutes, and no longer. Grabbing the apparatus when it is over 50 degrees Celsius may require gloves.
2. After the apparatus is shaken, keep it tilted upward. The vacuum line should be on top and the plate should be standing. When the cylinder valve is first opened the fluid that enters the apparatus can be in form of a vapor. Standing the apparatus up, gives the hot vapor a place to flow, instead of into the cell. Some of the vapor will fill the vacuum line.
3. Right before the valves are manuevered, the cell needs to be packed with ice. The ice should be in a very small bag, only big enough to cover the cell and not the apparatus. This should help reduce the chance of a large air bubble in the cell. Not all of the vapor that emerges from the cylinder, goes into the vacuum line. Some of it will enter the cell. If it hits the cell at one phase and then is cooled down immediately, by the ice, it may go to liquid. It doesn't matter if it also goes to two phase because it is already in the cell.

After the cell is sealed, disconnect the vacuum line at a place away from the apparatus. Leave the vacuum valve closed until the line is disconnected and the remaining pressure from the vacuum is relieved. If the cell is not sealed properly, and the vacuum valve is opened after filling, the residual vacuum may pull the fluid out of the cell. If the cylinder valve is not closed it may draw the fluid from there as well. Open the vacuum valve to allow the apparatus to dry out and cool.

When the apparatus is cool, remove the cell from the plate and apparatus. First, take out the secure screws that are holding the cell to the plate. Second, disconnect the cell between the 1/16" tubing and the c-a connector. Vise-grips placed gently on the hex of the c-a copnnector may transfer the force from the soldered joint.

Leak Testing the Cell: The Mass Method

The mass method of leak detection replaces the preferred procedure that involves a leak detector. The mass method is actually very simple, however, it is not as efficient as a leak detector. Throughout the process it is imperative that the cell is not handled. Fingerprints can hinder results when dealing with exact massings. The procedure is as follows:

1. When the cell is cool, mass and record its weight.
2. Place the cell in a vacuum oven at ~60°C and -20 psi overnight.

3. Remove the cell from the oven and allow to cool.
4. Mass the cell.
5. Repeat procedure.

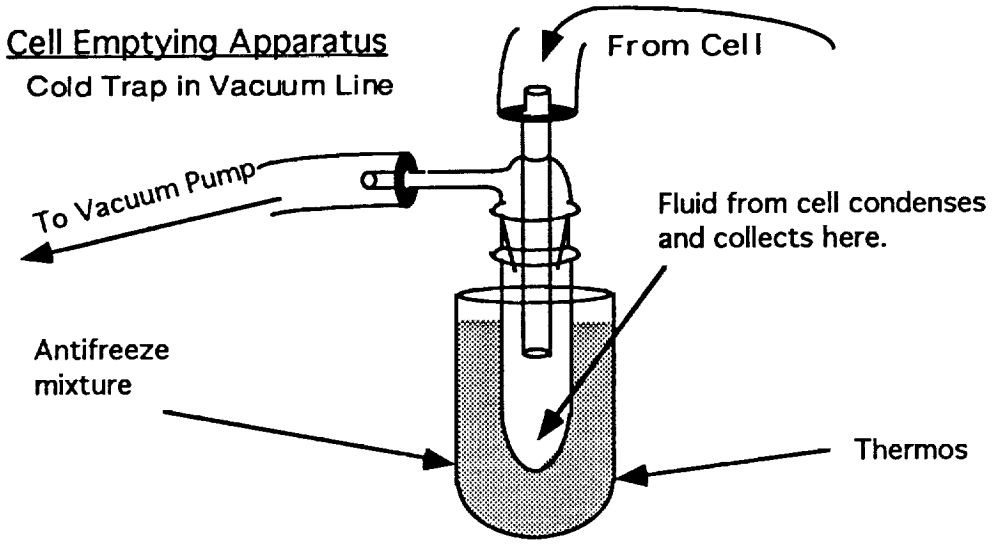
The cell may lose a small amount of weight because some fluids trapped in the threads of the c-a connector. However, it should not lose more than 1/100 g that way. Large losses in weight means the cell leaks. Repeating the procedure over and over will verify any assumptions.

If the cell leaks, empty it following the instructions below and attempt refilling. There are only two places the cell can leak. One is at the cell sealing screw which can be fixed by tightening it. The other place is at the knife edge. If it is simply a matter of having a copper knife edge that is not crushed enough, torque the support screws slowly to about 7 in.-lbs. using the same technique as was discussed on page 15.

Emptying Apparatus

It may be necessary to empty the cell for a number of reasons. The composition of the fluids may be too far from the critical composition or there may be a vary large vapor bubble in the cell. Maybe the cell was filled with methanol only to test the filling procedure and then needs to be emptied. Despite the reason, it may be helpful to know a simple technique that will empty the cell.

The apparatus uses the cold trap connected to a vacuum pump. The vacuum forces everything to be drawn through the cold trap. The cold trap sits in an antifreeze water mixture cooled with a cold finger. When the fluid enters the cold trap it condenses in the bottom of the trap and doesn't get drawn any further. This apparatus keeps fluids from being drawn through the vacuum pump, while also drawing a vacuum. It also keeps pump oil from contaminating the cell. See diagram below.



PART V. WATER BATH OBSERVATIONS/TESTING CRITICAL COMPOSITION

Testing the quality of the composition of methanol cyclohexane in the turbidity cell involves observing the fluids as they go from one phase to two phase and noting the temperature at which this occurs. Measuring the critical temperature, as this is called, is only one way to test the critical composition. Another way is by noting where the meniscus between the two fluids lies when they are two phase. The first method is more accurate than the second simply because it not only involves observation, it also involves temperature measurement related to phase transition, a much better indicator of critical composition. Both of these methods are, unfortunately, only as accurate as the apparatus involved. The first method is only as accurate as the probe that measures the temperature of the cell. The second relies on the visual appearance of a meniscus. The ability to tell where the meniscus lies depends on the shape of the cell and the quality of the fill. A vapor bubble in a round cell will make it hard to distinguish how much fluid is above the meniscus and how much is below. Due to time limitations, research done during 1992 involved only preliminary conclusions about the quality of the critical composition in each of two cells filled with methanol cyclohexane. These cells differed in that one had aluminum flanges and one had copper. Consequently, when mentioning the cells they shall be referred to as either the Al. cell or the Cu. cell.

Water Bath Set-up

The cell was mounted on a nylon rod and emerged in a insulated water bath. Temperature was controlled by two devices. An MGW Lauda RM 6 helped to stabilize the temperature and hold it up near 50 degrees Celsius. A Tronac PTC-41 stabilized the temperature further and provided precise temperature control of $\pm 0.0016^{o}C$.

Binary Fluid Meniscus Theory

Critical composition testing by measuring or estimating where the meniscus lies between the two fluids in two phase depends heavily upon the shape of the cell and quality of the fill. The cells used in the Summer of 1992 were round which means that a meniscus in the middle of the cell could have been easy to see, however, vapor bubbles in the cell lowered the amount of fluid. A vapor bubble at the top of the round cell caused the fluid to be curved which hindered seeing a meniscus and phase relationship. The Al. cell especially had a large vapor bubble in it. The Cu. cell has a much smaller bubble in it, however, due to improvements made in the filling procedure.

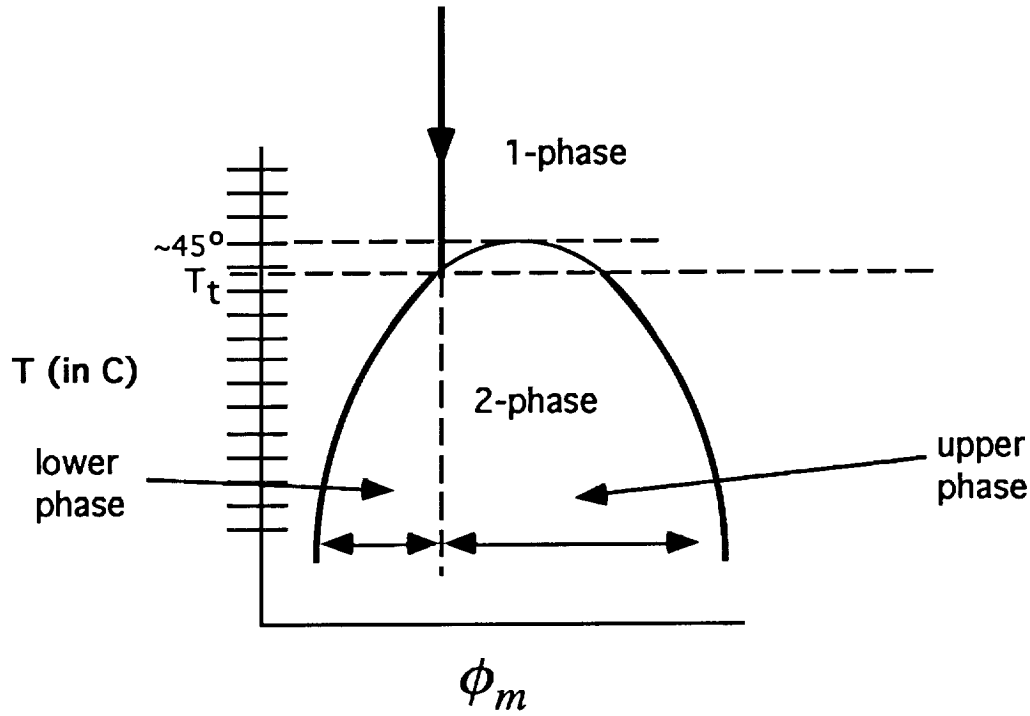
The theory behind the meniscus of the binary fluid mixture of methanol cyclohexane can be illustrated with the coexistence curve of the mixture. The illustration is on the next page. It is a graph of temperature verses the amount of methanol. If the sample is over it's transition temperature it is in one phase. As the temperature is dropped slowly, close the transition temperature it will begin to go to a two phase mixture and if allowed to settle a meniscus will form between the two phases. The phases are known as an upper and a lower phase. Dealing specifically with methanol cyclohexane, one phase will be a methanol rich phase and one will be a cyclohexane rich phase.

The arrow running down represents a sample of methanol cyclohexane. It's transition temperature seems to be a little under 44 degrees Celsius. At this point, a meniscus will form between the two fluids. The meniscus level is proportional to the lower and upper phases. This means that if the temperature is dropped slow enough, the meniscus will move slightly up or down, immediately after phase transition.

The critical composition of methanol cyclohexane has a transition temperature of about 45 degrees Celsius. Studying the graph, it is obvious, that if it is a critical

composition, the meniscus will not move at all, and it will move more the farther away from the critical composition the sample is.

One problem with this approach is that impurities can cause differing coexistence curves. The top of the curve could be at about 47 degrees or higher. Then the transition temperature doesn't help determine if the sample is of critical composition.



Water Bath Techniques

The following technique was found to be the most efficient when trying to narrow the critical point of the mixtures down. The bath was first taken to a temperature where the cell was certain to be in one phase. Then the temperature was dropped until the cell went to two phase. The first indication that a cell or mixture is changing is a cloudy appearance. This critical opalescence occurs very close to the critical point. The next step is to take the cell into one phase by raising the temperature about half way to where the cell is known to be in one phase. When the temperature is steady there, it is important that the cell is agitated to see if it will stay in one phase or return to two. If it returns to two phase, with the continued onset of a cloudy appearance, then raise the temperature half again of how far it is from the initial temperature and wait for the temperature to steady. If the cell goes to one phase after agitation, then lower it half way down to where it is known to be in two phase. Continuing this procedure will quickly narrow the temperature down close to the critical temperature.

Results

While this project was to concentrate on filling a cell with a critical mixture of Methanol-Cyclohexane, it soon became evident that finding a way to seal the cell would be a major challenge. In addition to redesigning the fill line connection, numerous other attempts were made to rework the cell to keep it sealed. Different spacers were tried and some spacers were remachined at the University of Maryland's Space Science Shop to improve the copper knife-edge. Many of these would seal at room temperature but would fail catastrophically (the fluids would all be gone) after the cell was heated and put under a vacuum. In an effort to minimize thermal expansion effects that might cause the cell failure upon temperature cycling, different support screws were tried including stainless steel, titanium and specially-made Invar bolts. All gave similar results. Another cell sealing technique used an Indium ring to seal between the copper spacer and the window. However, Indium is too soft and while better and easier seals could be made at room temperature, these cells also failed when temperature cycling occurred. While we felt the leaks occurred at the knife-edges, we could never rule out the cell sealing screw. It was also noticed that the Aluminum flange cells were harder to seal and the windows cracked more often, in part due to the ease with which the flanges could deform and cause large local forces on the windows.

A number of positive features resulted from the summer's research that will be important to the continuation of this research (as NASA grant NAG3-1404).

1) A cell fill assembly capable of transferring a critical fluid mixture in the one phase region at elevated temperatures was demonstrated successfully.

2) A filled cell could be tested to determine if the critical composition was present and, if not, how far off critical it might be.

3) The present cell design was found to be inadequate for this experiment and a new design will be needed for the subsequent experiment. Either a similar sandwich design with a gold o-ring will be used or a completely redesigned cell will be done.

The students (James Becker and Anne Flewelling) learned a number of experimental techniques in this project. Together we have struggled with the difficulties associated with experimental research which probes the frontier in a field. Our experience has provided a number of insights and knowledge in designing cells of this type and confidence in a future cell design.

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